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Solving a 9σ discrepancy between hyperfine theory and experiment in trapped HD^+ ions

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Previously we have carried out Doppler-free laser vibrational spectroscopy of trapped, laser-cooled HD^+ molecular ions with a relative uncertainty of a few parts per trillion (ppt) [1]. Combined with accurate theoretical predictions and other recent precision measurements, our HD^+ data can potentially improve the literature value of the electron's relative atomic mass from 29 ppt to 18 ppt [2]. Surprisingly, the Doppler-free spectroscopy also revealed a large (8.5 kHz, or 9σ) deviation between the observed and theoretically predicted hyperfine structure. In order to resolve the 9σ discrepancy, we are currently performing electron spin resonance spectroscopy of various hyperfine transitions in HD^+ to measure the hyperfine structure with a target uncertainty of 0.1 kHz. The results should allow establishing whether the discrepancy stems from proton-electron, deuteron-electron, or spin-rotation interactions, and/or from an extraordinarily large yet overlooked systematic effect in the previous experiments.

[1] Patra et al., *Science* **369**, 1238-1241 (2020)

[2] Karr and Koelemeij, *Mol. Phys.* (2023) DOI: 10.1080/00268976.2023.2216081

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